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SOME PROBLEMS RELATIVE TO CALCULATION AND MEASUREMENT OF MICROWAVE AND SUBMICROWAVE ABSORPTION IN ATMOSPHERIC WATER VAPORS

by

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SOME PROBLEMS RELATIVE TO CALCULATION AND MEASUREMENT OF MICROWAVE AND SUBMICROWAVE ABSORPTION IN ATMOSPHERIC WATER VAPORS *

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by S. A. Zhevakin
& A. P. Naumov

SUMMARY

It is noted that the latest measurements of the absorption coefficient of water vapor in the relative transparency windows of the submicrowaves ($\lambda > 60\mu$), performed by a series of authors, exceed the earlier values computed by us by $\sim 1.5 \div 2$ times, that is, the same departure takes place between theory and experiment as was found in the microwave band.

The possible causes of such a discrepancy are discussed alongside with the possibilities of experimental investigation of submicrowave absorption by radioastronomical methods.

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* *

The methods and the results of quanto-mechanical calculation of the absorption coefficient γ of water vapors in the atmosphere for the wave band $\lambda = 10\mu \div 32$ cm were expounded in detail in the works [1 — 3] **. Comparison between the experimental and theoretical results of obtaining the absorption factor was also drawn in the above-mentioned works. A reliably established excess by $\sim 1.5 \div 2$ times of the measured values of γ over the theoretical ones in the microwave region of the spectrum $2\text{ mm} < \lambda < 8\text{ mm}$ was demonstrated in particular [1, 3]. As to the submicrowave band, there was no possibility to derive in [1] reliable conclusions about the degree of correspondence between absorption theory and experiment, which was due to insufficient measurements. Since then a series of experimental of radiowave absorption by water vapors in the region $\lambda > 60\mu$ were completed [4 — 7].

* NEKOTORYYE VOPROSY RASCHETA I IZMERENIY POGLOSHCHENIYA MILLIMETROVYKH I SUBMILLIMETROVYKH RADIOVOLN V ATOSFERNYKH PARAKH VODY.

** [See ST-RWP-AI-10354 and also 209].

Now we have evidence that in the submicrowave band, beyond the spectral lines of H_2O, the values computed in [1] for the absorption coefficients of water vapor are less than the experimental ones by about the same amount, that is, $1.5 \div 2$ times.

The insufficient resolution of the apparatus utilized in the works [4 — 7] did not allow the authors of these works to measure the absorption coefficient in the spectral lines of water vapor. Meanwhile such measurements might have contributedin throwing light on the possible causes of discrepancies bwteeen the theory of absorpion in water vapor and the experiments in relative transparency windows. The astronomical method (see [5]) of determination of γ by the relative variation of solar radiation, weakened by the atmosphere, for various heights of the Sun above horizon may be utilized only on conditions of such radiowave absorption in the atmosphere, when the Sun's antenna temperature in the given frequency band is sufficiently high and varies notably with the zenithal angle change. Since the absorption coefficient of water vapor γ exceeds by tens and hundreds of times the resonance values of γ in centimeter and microwaves even in the transparency windows of submicrowaves, the radioastronomical methods of measurement of the absorption coefficient (by Sun's and atmosphere's proper radiation) have a very limited area of applicability in the region $\lambda < 1$ mm. In order to corroborate the conclusion derived, we shall briefly pause at the quantitative characteristics of the effective Sun's and atmosphere's radiation in the submicrowave band, and at conclusion we shall discuss the possible causes of the above-noted discrepancy of water vapor absorption theory with the experiment in these waves.

1.- We plotted in Figures 1 and 2 the effective temperature of solar radiation, $T_C = T_{\odot} e^{-\tau}$, weakened in the terrestrial atmosphere, but without taking into account the atmosphere's proper emission (about the relative role of the latter, see below). The computations of the quantity T_C were performed in the band $1/\lambda = 5 \div 35$ cm^{-1} ($\lambda \simeq 0.28 \div 2$ mm) at various zenithal angles θ, for all the observations conducted from a height $h \simeq 4$ km (pressure $P = 478$ mm Hg, near-Earth absolute moisture $\rho = 1$ and 2 g$\cdot$m^{-3}), that is, for conditions close to those having taken place at measurements [5]. At the same time, it was assumed at either the calculation of T_C

(see Figs 1 and 2), of $T_{эфф}$ (Figs 4 and 5), that is, the effective temperature of the atmosphere, that the absorption of radiowaves $\lambda \leq 2$ mm is determined only by water vapor. It may be seen from Fig. 2 of the work [30] and the Fig. 3 of the present work that at sea level, where the absolute humidity is $\rho = 7.5$ g·cm^{-3} the ratio of the absorption coefficient of molecular oxygen to that of water vapor is equal to $\simeq 0.04$ even at $\lambda \simeq 1 \div 2$ mm (in the wavelength $\lambda \simeq 0.87$ mm, that is, the nearest from the side of submicrowaves transparency window) this ratio still decreases by $\simeq$ 10 times), the assumption made is still fulfilled.

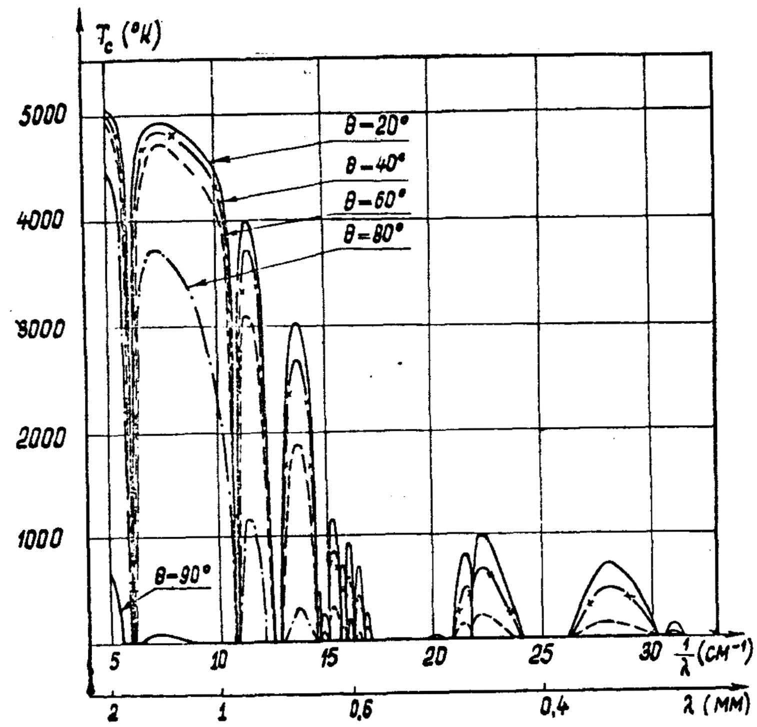

Fig. 1. - Temperature of atmosphere-weakened solar radiation T_C (without taking into account the atmosphere's proper radiation) at $h \simeq 4$ km (pressure $P = 478$ mm Hg, absolute humidity $\rho = 1$g.m^{-3}) for various values of zenithal angles θ. The effective temperature of the Sun $T_{\odot}$ in the band $\lambda = 0.28 \div 2$ mm ($1/\lambda = 5 \div 35$ cm^{-1}) is taken equal to 5 200° K; temperature of the atmosphere $T_A = 283$°K.

Utilizing the standard transformation rules, we have for the ratio of absorption coefficients O_2 to H_2O at $h \simeq 4$ km, $\rho = 1$g·m^{-3} (see [1, 2, 30]) $\simeq 0.1$. However, according to measurements of [31] (see their processing in [32]), the oxygen absorption at $\lambda \simeq 1 \div 2$ mm, is about by one order higher than the computed data. The discrepancy for the case of O_2 absorption factors for $\lambda \leqslant 2$ mm may be caused by an inaccurate accounting of nonresonance absorption of O_2 in the calculation of the work [30]. If the subsequent experimental investigations should confirm such a high value of the absorption coefficient of O_2 ($\gamma_{O_2} \simeq 0.17$ db/km) at sea level with $\lambda \simeq 1 \div 2$ mm, at $h \simeq 4$ km, the absorption coefficients of oxygen and water vapor at $\lambda \simeq 1 \div 2$ mm and $\rho = 1\,g{\cdot}m^{-3}$ would be mutually comparable.

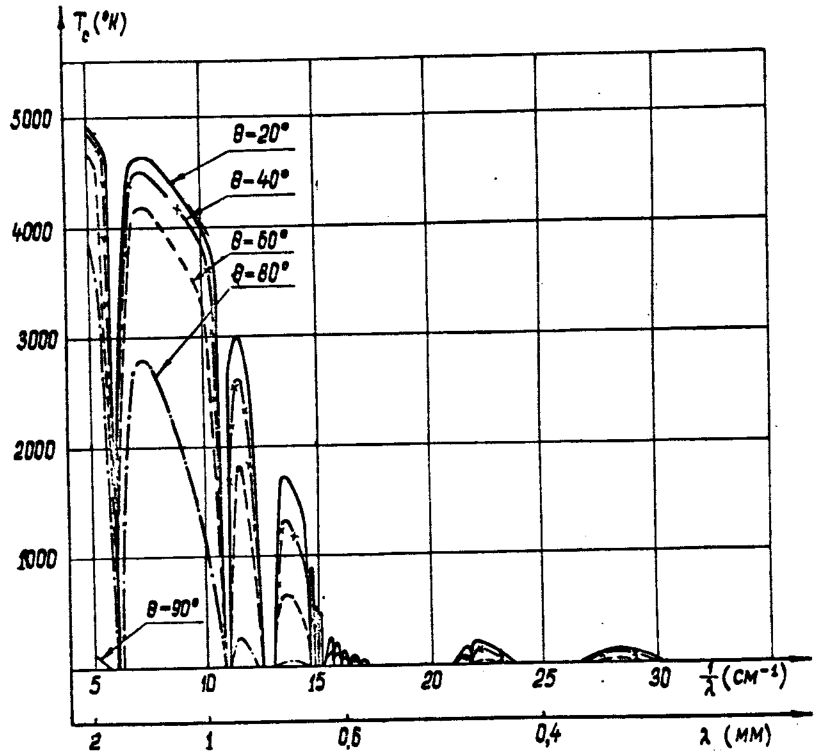

Fig. 2. - Temperature of solar radiation T_C, weakened by the atmosphere (without taking into account its proper emission) at $h \simeq 4$ km (pressure $P = 478$ mm Hg, absolute moisture $2\,g{\cdot}m^{-3}$) for the values of zenithal angles $\theta = 20^o, 40^o, 60^o, 80^o, 90^o$. The effective temperature of the Sun $T_{\odot}$ in the band $\lambda = 28 \div 2$mm ($1/\lambda = 5 \div 35\,cm^{-1}$) is taken at 5200^oK; atm.temp. $T_A = 283^o$ K.

The accounting of that circumstance will lead in its turn to a decrease in the computed values of T_C (see Fig. 1) in the band $1/\lambda \simeq \simeq 6.3 \div 10\,cm^{-1}$ ($\simeq 1 \div 1.6$ mm) at $\theta = 20 \div 60^o$ by no more than $\simeq 25\%$ and to an increase of $T_{эфф}$ in the same band but with $\theta = 20 \div 80^o$ (see Fig. 4) by $\simeq 1.8$ times (at $\rho = 2\,g{\cdot}m^{-3}$ the respective corrections will be $\simeq 12\%$ for T_C (Fig.2) and $\simeq 1.3$ times for $T_{эфф}$ (Fig. 5)).

However, in submicrowaves ($\lambda \leqslant 0.9$ mm), the results of the calculation, plotted in Figs. 1, 2, 4, 5, are practically unchanged on account of the absorption by water vapor clearly prevailing here (the error should not be higher than tenths of fractions of percent).

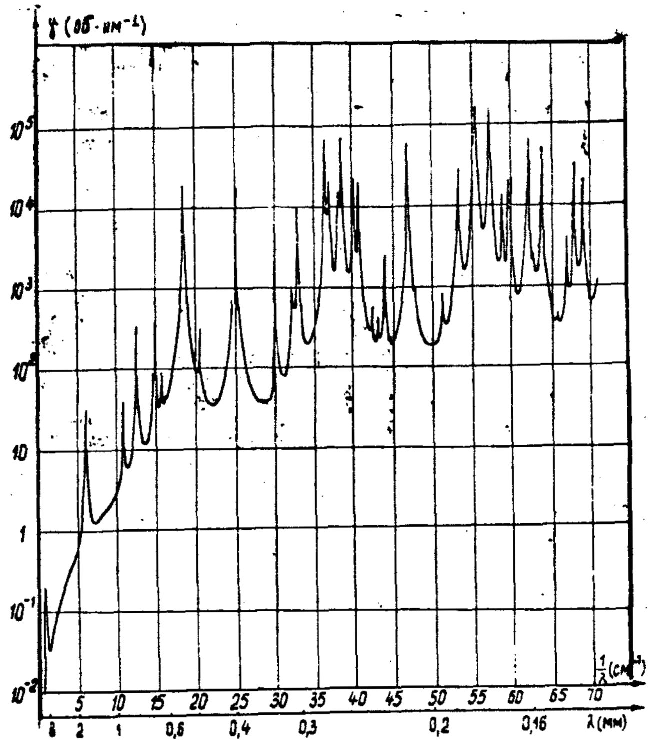

Fig. 3. - Absorption coefficient γ of the atmospheric water vapor in the band $\lambda \simeq 0.14$ mm $\div$ 2 cm ($1/\lambda = 0.5 \div 70$ cm^{-1}) at the absolute moisture $\rho = 7.5\, g \cdot m^{-3}$, temperature $T_A = 293^\circ$ K and pressure $P = 760$ mm Hg.

When computing $T_C = T_{\odot} e^{-\tau}$ (the Rayleigh-Jeans approximation), it was assumed that the effective temperature of the Sun is $T_{\odot} = 5200^\circ$ K in the entire band $\lambda = 0.28 \div 2$ mm and the opacity is $\tau = \bar{\gamma} l_2$ where $\bar{\gamma}$ is the absorption coefficient of water vapor at $h = 4$ km, expressed in nepers, and l_2 is the effective path length of radiowaves in water vapor, reduced to the conditions on the Earth's surface. The quantity l_2 is found taking into account the radiowave refraction according to the method described in [8]. The only exception consisted in that we utilized the characteristic height of the absorption coefficient of water vapor H_{H_2O}, equal to 1.5 km [9], instead of 2.1 km, as was done in [8], the latter's seasonal variations being $\leqslant 5\%$ [9].

Beyond the resonance frequencies of water vapors, that is, in the most interesting spectral regions,

$$\bar{\gamma} = \frac{1}{10\log_{10} e}\gamma(0)\frac{P}{760}\frac{\rho}{7,5}\frac{293}{T_A}$$

where $\gamma(0)$ is the absorption coefficient of water vapor at sea level, at $\rho = 7.5\,g\cdot m^{-3}$, and expressed in $db\cdot km^{-1}$ (the temperature dependence $\bar{\gamma} \sim 1/T_A$ takes place in relative transparency windows at $\lambda \geqslant 0.2$ mm [2]). In the above calculation we utilized the values $\gamma(0) = 1.5\,\gamma$, where γ is the value of the absorption coefficient at sea level at moisture $\rho = 7.5\,g\,m^{-3}$, obtained in [1] (see Fig. 3). The introduction of the multiplier 1.5 everywhere,

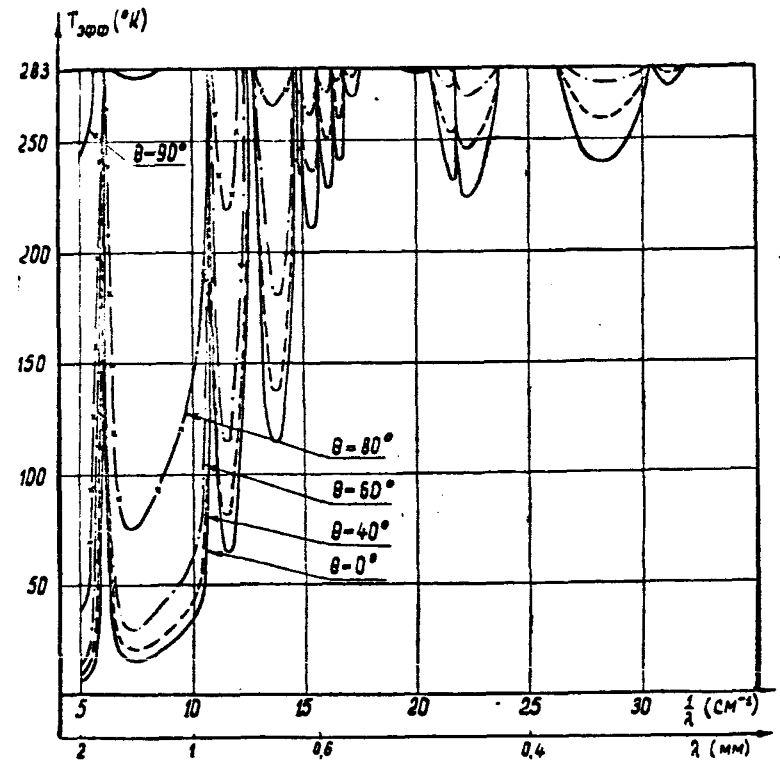

Fig. 4.- Effective temperature of atmospheric radiation $T_{эфф}$ at $h \simeq 4$ km (pressure $P = 478$ mm Hg, absolute moisture $\rho = 1\,g\cdot m^{-3}$; temperature $T_A = 283°$ K) in the band $\simeq 0.28 \div 2$ mm ($1/\lambda = 5 \div 35\ cm^{-1}$) for various values of the zenithal angle θ.

except for the narrow frequency intervals in resonances of the order of the width of spectral lines $2\Delta\nu/c \simeq 0,2\ cm^{-1}$ ($\Delta\nu$ being the half-width of the spectral line in cps and c — the speed of light) was made for the elimination of the above-noted discrepancy between the computed and the measured values of the absorption coefficient.

Here we make use of the case and in Fig. 3 we bring forth anew the dependence of the absorption coefficient of water vapor γ on wavelength in the $\lambda \simeq 0.14\,\text{mm} \div 2\,\text{cm}$ at sea level, for when computing the quantity γ in [1, 2], an error was admitted in the intensities of spectral lines $9_8 - 10_6$ and $9_9 - 10_5$ $(1/\lambda_{ij} = 28.99\ \text{cm}^{-1}$; λ_{ij} is the resonance wavelength); this error led in the resonance to the value $\gamma \simeq 5000\ \text{db/km}^{-1}$ instead $\gamma \simeq 40\ \text{db/km}^{-1}$, which did not conform with the measurements made in [6, 7, 10] (cf Fig. 3 of this work with Fig. 1 in [1]). The calculations performed now are in complete qualitative agreement with the measurements of [4 — 7, 10]. The error noted affects the calculations of the absorption coefficients of water vapors only in a narrow spectral band $1/\lambda \simeq 27 \div 30.2\ \text{cm}^{-1}$ $\lambda \simeq 0.33 \div 0.37\,\text{mm}$) and does not modify at all (discrepancy $< 0.1\%$) the calculated results outside the indicated band (see Figs 1 — 15 in [1] and Figs 2 — 13 in [2]).

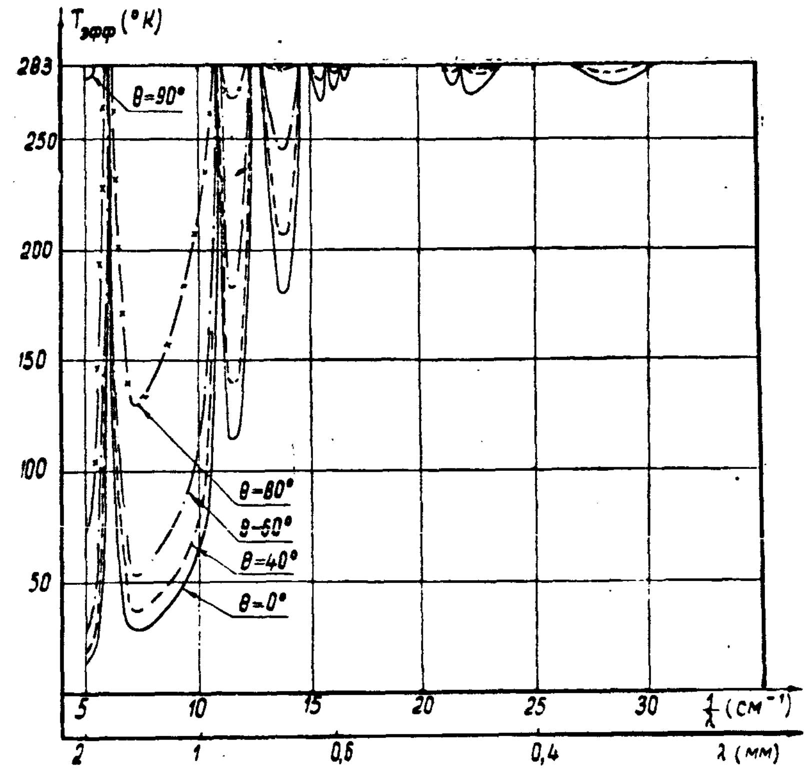

Fig. 5. - Effective temperature of atmospheric radiation $T_{эф}$ at $h \simeq 4\,\text{km}$ (pressure $P = 478\,\text{mm Hg}$, absolute moisture $\rho = 2\,\text{g m}^{-3}$ temperature $T_A = 283°\,\text{K}$) for various values of zenithal angles θ in the band $\lambda \simeq 0.28 \div 2\,\text{mm}$ $(1/\lambda = 5 \div 35\,\text{cm}^{-1})$.

For the computation of T_C we utilized the value $T_{\odot} = 5200^\circ$ K, which is obtained at interpolation of the results of measurements of Sun's effective remperature in the infrared and microwave bands (see the review of publications in the #8 of the monograph [11], and also the works [12 - 16] (the measurements of $T_{\odot}$ at the neighboring submicrowaves were not made). If $T_{\odot}$ is subsequently made more precise in submicrowaves, it will be easier for us to introduce in our computed value of T_C the proper correction, for T_C is directly proportional to $T_{\odot}$. It is, however, difficult to expect in any case that the chosen value of $T_{\odot}$ differs by more than 10% from the true value, and consequently, the possibilities of radioastronomical investigations in submicrowave band may be estimated with enough precision by the Figs. 1 and 2.

It follows from these two figures, in particular, that in relative transparency windows of submicrowaves $1/\lambda \simeq 11.5$ cm^{-1} ($\lambda \simeq 0.87$ mm); $1/\lambda \simeq 13.5$ cm^{-1} ($\lambda \simeq 0.74$ mm) and $h = 4$ km the difference of solar temperatures ΔT_C, taking into account the atmospheric weakening for various zenithal angles θ ($\theta = 20 \div 60^\circ$) and $\rho = 2$ g m^{-3} constitutes $\simeq 400 \div 500^\circ$ K at $\Delta\theta = 20^\circ$. Such a difference in radiation temperatures may be registered by contemporary indicators, which allows to apply successfully the radioastronomical method of measurement of atmospheric absorption by the Sun in these transparency windows (the accounting of atmosphere's proper radiation may increase the difference thus found by no more than $\sim 30^\circ$ K, as may be seen from Figs. 4 and 5).

It may also be seen from Fig. 2 that at $\rho = 2$ g m^{-3}, at $h = 4$ km and at wavelengths $\lambda < 0.6$ mm the solar radiation is practically entirely absorbed in the atmosphere column lying above the indicated height. This absorption takes place at $\gamma \gtrsim 9$ db km^{-1} and $h = 4$ km, which corresponds to sea level at $\rho = 7.5$ g m^{-3} and $\lambda \gtrsim 60$ db km^{-1}. To such values of γ respond waves in the band $27.4\,\mu < \lambda < 600\,\mu$ (see Fig. 3 in the present work and Figs 2 — 15 in [1]).

As the absolute moisture increases the quantity ΔT_C decreases rapidly in the transparency windows of water vapor. Thus, the calculations performed for normal atmosphere conditions ($P = 760$ mm Hg, $\rho = 7.5$ g m^{-3}), attest to the impossibility of absorption measurements by the weakening of solar radiation from sea level even in the transparency window $1/\lambda \simeq 11.5$ cm^{-1} ($\lambda \simeq 87$ mm). As to the effective temperature of atmosphere radiation $T_{эфф} = T_A(1 - e^{-\tau})$, the difference $\Delta T_{эфф}$, corresponding to $\Delta\theta = 40°$, it does not exceed $\simeq 100°$ K in wavelengths $\lambda = 0.74$ mm and $\lambda = 87$ mm (Fig. 4, 5) (the estimates were performed for an isothermic atmosphere. The correction for nonisothermicity of the atmosphere affects the results by no more than $\simeq 10°$ K. Consequently, absorption measurements by solar radiation in the near submicrowave band are obviously preferable to measurements by atmosphere's proper radiation, though the first method may also be utilized, but only in special conditions.

2.- The possible causes of discrepancies between experiment and theory of micro and centimeter wave absorption in water vapor were more than once discussed in the press [3]. It is not excluded that the undisclosed and unaccounted for factors of microwave absorption, leading to such discrepancies, may also be essential on near submicrowaves. It was shown in [3] that the theoretical values of the absorption coefficient of water vapors may be drawn nearer the experimental by applying in the calculation a line shape obtained from the solution of the kinetic equation instead of the Van Vleck-Weiskopf line *. However it is then practically impossible to attain a full accord between theory and experiment in the $2\,mm < \lambda < 8\,mm$ region. In submicrowaves the difference in the shape of spectral lines manifests itself in transparency windows only in the nearest region ($\lambda = 0.74$ mm, $\lambda = 0.87$ mm) and at $\lambda < 27\mu$ (see Fig. 1 – 15 in [1]). However, in the $27\mu < \lambda < 600\mu$ band calculations lead to nearly identical results with lines of different shapes. The absence of of measurements of the absorption coefficient of submicrowaves in all

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* In the structural factor for the kinetic equation, a misprint has been admitted in [1], where formula (7) should be read as :

$$f(\nu, \nu_{ij}) = \frac{\nu^2}{\pi} \cdot \frac{4\Delta\nu_{ij}}{(\nu_{ij}^2 - \nu^2)^2 + 4\nu^2(\Delta\nu_{ij})^2}.$$

** Nearly in all the geophysical computations linked with molecular absorption [17, 18], the Lorenz line shape is applied. Criticism of the original Lorenz model can be found in [19, 20]. However, the dispersion Lorenz correlation describes satisfactorily the infrared spectra of atmospheric gases

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resonance frequencies of water vapors hinders the analysis of causes of discrepancies between theory and experiment in transparency windows.

In reality, is the excess of experimental absorption coefficients over theroretical due to additional absorption factors, for example polymeric formations of H_2O molecules [21]*, or is it a consequence of any inaccuracies in the description of the rotational spectrum of H_2O monomers, as for example the imprecise value of spectral line widths, or is this discrepancy linked with a simultaneous presence of a series of causes ? This will become clearer in case of expansion of the area of experimental investigations.

Some doubts have been lately voiced as to the exactitude of the half-widths of water vapors' spectral lines computed by Benedict and Kaplan [22, 23], using the Anderson theory [24]; they were utilized by us in the computations [1 – 3]. It is well known that the main contribution to the widening of rotational lines of water vapor in the atmosphere is made by molecular collisions $H_2O — N_2$ and the half-width of the line (quantum transition $i — j$) in the air $(\Delta\nu/c)_{ij}^{H_2O-air}$ constitutes about 90% of the corresponding quantity $(\Delta\nu/c)_{ij}^{H_2O-N_2}$ [1]. The detailed calculations of water vapor line widening for the collisions $H_2O - H_2O$ and $H_2O - O_2$, performed in [25], confirm the validity of that estimate. However, when computing the quantities $(\Delta\nu/c)_{ij}^{H_2O-N_2}$ Benedict and Kaplan [23] had strictly taken into account only the dipole-quadrupole interaction of molecules $H_2O — N_2$. The latter, alongside with the closely-acting interaction (quadrupole-quadrupole, dipole-induced dipole, and so forth) determines the collision parameter b_m, equal to the distance between colliding molecules at their closest rapprochement. The value of b_m and the quadrupole moment of the nitrogen molecule have been assorted in [23] from the comparison of the thus computed and experimental values of half-widths of the water vapor line $5_{-1} — 6_{-5}$ (ij — 1.35 cm). Therefore, all the half -widths of the lines $(\Delta\nu/c)_{ij}^{H_2O-N_2}$, computed in [23], are normalized to the measured value $(\Delta\nu/c)_{ij}^{H_2O-air}$ for the transition $5_{-1} — 6_{-5}$.

.. (continued from the preceding page..) at least in regions, distant from resonance by a few line widths. It is interesting to note that the absorption coefficient of water vapor in the band $\lambda \simeq 10\mu$ - 32 cm, computed by us with the Lorenz line shape, differs from the results [1] obtained with line shape by kinetic equation by no more than 5%. Insamuch as the latter strictly follows at specific idealizations, the just noted result coincidence allows also the use of the Lorenz structural factor as a fortunate approximation of the shape of the spectral line.

It is noted in [22] that the half-width of three intense water vapor lines ($1_1 - 2_1$, $1/\lambda_{ij} = 92{,}54$ см$^{-1}$; $3_1 - 4_1$, $1/\lambda_{ij} = 170{,}33$ см$^{-1}$; $4_0 - 5_0$, $1/\lambda_{ij} = 188{,}27$ см$^{-1}$), apparently exceed the corresponding values given by Benedict and Kaplan, by $\sim 1.5 \div 1.8$ times. If the widths of other spectral lines too were by as many times greater than the computed ones (if only the most intense ones), the theoretical [1 — 3] and experimental [4 – 7] absorption coefficients of water vapor would be in total quantitative agreement, for in relative transparency windows there takes place the correlation $\gamma \sim (\Delta\nu/c)_{ij}^{H_2O-N_2}$ (see formula (15) in [1]). If the contribution of the given line exceeds a great deal the aggregate contribution of all the remaining spectral lines, then in the resonance frequency $\gamma \sim 1/(\Delta\nu/c)_{ij}^{H_2O-N_2}$, that is, when the refining of the widths of spectral lines is possible, the data on absorption by water vapors [1 – 3] in the peaks of spectral lines are easy to reduce to new values of $(\Delta\nu/c)_{ij}^{H_2O-N_2}$.

On the other hand, it should be pointed out that the widths of water vapor lines were not measured directly in [22], and they were only estimated by the magnitude of integral absorption in a certain band. Meanwhile, spectroscopic measurements of the half-width of the water vapor line $2_2 - 3_{-2}$ ($1/\lambda_{ij} = 6.12$ cm^{-1}) [29] gave in the air $(\Delta\nu/C)_{exper} = 0.086$ cm^{-1} as against the theoretical value of Benedict & Kaplan $(\Delta\nu/C)_{theor} = 0.091$ cm^{-1}, that is, the discrepancy for the width of the indicated line constitutes only $\simeq 6\%$. If in addition to that the coincidence of computed and measured absorption coefficients of water vapor in the band $\lambda \simeq 10\mu$ is considered as indirect evidence of the reliability of lines' H_2O half-widths computed by Benedikt & Kaplan [23], the search for additional agents of absorption, whose spectrum is localized on long submicro and microwaves, should be approached with considerably greater attention. One may think indeed, that the correspondence existing at $\lambda \approx 10\mu$ between theory and experiment is attained on account of inaccuracies in the other para eters of the H_2O spectrum. But the other possible inaccuracies (description of absorption in the wings of spectral lines, the nonhardness effect of the H_2O molecule on line intensity, the contribution of H_2O isotopes to absorption) were already the object of discussions in [1, 3]. It is noted in these works that all the circumstances brought forth above cannot apparently modify the computed absorption coefficient of water vapors by more than 10% at $\lambda > 100\mu$ and 25% at $\lambda \lesssim 100\mu$.

Let us remark, in conclusion, that certain questions of atmosphere physics and the requirement of a detailed clarification of the propagation conditions of microwaves in the atmosphere make prerequisite the knowledge of seasonal variations of absorption at different heights. This question is discussed at length in the work [9].

*** THE END ***

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